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et for filing a PROVISIONAL APPLICATION FOR PATENT under 37 CFR 1.53(c).

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Stanley	Leshaw	1= 14		7 Yorktown Road, Wayne NJ 07470		300	
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Additional inventors are being	ing named on thi separe	ately numbe	red sheets attached h	nereto		N.	
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## UV STABILIZING ADDITIVE COMPOSITION

#### FIELD OF THE INVENTION

This invention relates to an improved UV stabilizing additive composition. More specifically, this invention relates to an UV stabilizing additive composition comprising an ortho-hydroxy triazine compound, a hindered amine compound and hydroxybenzophenone compound.

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## **BACKGROUND OF THE INVENTION**

Exposure to sunlight and other sources of ultraviolet (UV) radiation is known to cause degradation of a wide variety of materials, especially polymeric materials. For example, polymeric materials such as plastics often discolor, lose gloss and/or become brittle as a result of prolonged exposure to UV light due primarily to a reduction in the molecular weight of the polymer. Accordingly, a large body of art has been developed directed towards materials such as UV light absorbers and stabilizers, which are capable of inhibiting such degradation in polymeric articles.

The present inventors have found that a combination of ortho-hydroxy triazine compound, a hindered amine compound and hydroxybenzophenone compound provides synergistic protection of materials against UV light. This combination either provides better protection at typical UV stabilizer loading levels, or typical protection at much lower loading levels resulting in significant cost savings over prior art UV stabilizing additive compositions.

#### SUMMARY OF THE INVENTION

This invention relates to an UV stabilizing additive composition comprising an orthohydroxy triazine compound, a hindered amine compound and a hydroxybenzophenone compound. This additive composition may be used to stabilize materials from UV radiation. This invention also contemplates a method of stabilizing a material by contacting the material with the UV stabilizing additive composition.

### **DETAILED DESCRIPTION OF THE INVENTION**

This invention relates to an UV stabilizing additive composition comprising an orthohydroxy triazine compound, a hindered amine light stabilizing (HALS) compound and a hydroxybenzophenone compound. Pr ferably, the ortho-hydroxy tris-aryl-s-triazine compound has the following formula

I:
$$R_{1} \longrightarrow (A) \longrightarrow (C) \longrightarrow R_{7}$$

$$R_{3} \longrightarrow (N) \longrightarrow (N) \longrightarrow (C) \longrightarrow (R_{7} \longrightarrow (R_{9} \longrightarrow (R_{9}$$

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where A, B and C are each aromatic, at least one of A, B and C is substituted by a hydroxy group ortho to the point of attachment to the triazine ring, and each of  $R_1$  through  $R_9$  is selected from the group consisting of hydrogen, hydroxy, alkyl, alkoxy, sulfonic, carboxy, halo, haloalkyl and acylamino having from about 1 to about 24 carbon atoms.

One embodiment of the tris-aryl-s-triazine is a compound having the formula II:

$$Ar_1 \longrightarrow N \longrightarrow R_{21}$$

where Ar<sub>1</sub> and Ar<sub>2</sub> are the same or different and are substituted or unsubstituted aryl groups; and where R<sub>20</sub> and R<sub>21</sub> are each independently a hydrogen, C<sub>1</sub>-C<sub>24</sub> alkyl, C<sub>1</sub>-C<sub>24</sub> haloalkyl, C<sub>6</sub>-C<sub>24</sub> aryl, C<sub>2</sub>-C<sub>24</sub> alkenyl, C<sub>1</sub>-C<sub>24</sub> acyl, C<sub>1</sub>-C<sub>24</sub> cycloalkyl, C<sub>5</sub>-C<sub>24</sub> cycloacyl, C<sub>7</sub>-C<sub>24</sub> aralkyl, or C<sub>6</sub>-C<sub>24</sub> aracyl, substituted or unsubstituted biphenylene, substituted or unsubstituted napthalene, OR, NRR', CONRR', OCOR, CN, SR, SO<sub>2</sub>R, and where R and R' are each independently a hydrogen, C<sub>1</sub>-C<sub>24</sub> alkyl, C<sub>1</sub>-C<sub>24</sub> haloalkyl, C<sub>6</sub>-C<sub>24</sub> aryl, C<sub>2</sub>-C<sub>24</sub> alkenyl, C<sub>1</sub>-C<sub>24</sub> acyl, C<sub>1</sub>-C<sub>24</sub> cycloalkyl, C<sub>5</sub>-C<sub>24</sub> cycloacyl, C<sub>7</sub>-C<sub>24</sub> aralkyl, or C<sub>6</sub>-C<sub>24</sub> aracyl, substituted or unsubstituted biphenylene, or substituted or unsubstituted napthalene.

Preferably, in the above formula II, Ar<sub>1</sub> has the formula IIa:

where  $R_{22}$  and  $R_{23}$  are each independently a hydrogen,  $C_1$ - $C_{24}$  alkyl,  $C_1$ - $C_{24}$  haloalkyl,  $C_6$ - $C_{24}$  aryl,  $C_2$ - $C_{24}$  alkenyl,  $C_1$ - $C_{24}$  acyl,  $C_1$ - $C_{24}$  cycloalkyl,  $C_5$ - $C_{24}$  cycloacyl,  $C_7$ - $C_{24}$  aralkyl, or  $C_6$ - $C_{24}$  aracyl, substituted or unsubstituted biphenylene, substituted or unsubstituted napthalene,  $C_1$ - $C_2$ - $C_3$ - $C_4$ - $C_5$ - $C_5$ - $C_6$ -C

Also preferred in the above formula II is when  $R_{20}$  is hydrogen or a  $C_1$ - $C_8$  alkyl,  $R_{21}$  is hydrogen and  $Ar_1$  and  $Ar_2$  may be the same or different and are benzyl, methylbenzyl, or dimethylbenzyl.

Examples of suitable tris-aryl-s-triazines that may be used are 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-n-octyloxyphenyl)-4,6-bis(2,4dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-(mixed iso-octyloxyphenyl)-4,6-bis(2,4dimethylphenyl)-1,3,5-triazine; 2-(2,4-dihydroxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5triazine; 2,4-bis(2-hydroxy-4-propyloxyphenyl)-6-(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[2-hydroxy-4-(2-hydroxy-3butyloxypropyloxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[2-hydroxy-4-(2hydroxy-3-octyloxypropyloxy)-phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[4dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxyphenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxypropoxy)phenyl]-4,6-bis(2,4dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5-triazine; 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine; 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxypropoxy)phenyl]-1,3,5-triazine and 2-(2-hydroxyphenyl)-4-(4-methoxyphenyl)-6phenyl-1,3,5-triazine.

The hindered amine compound may be any suitable hindered amine compound such as those containing a 2,2,6,6-tetraalkylpiperdine or 2,2,6,6-tetraalkylpiperazinone radical. One embodiment of a hindered amine compound is one that contains at least one group having the following formula III:

$$\begin{array}{c|c} & & & & \\ & & & \\ R_{15} & & & \\ & & & \\ R_{16} & & & \\ & & & \\ R_{14} & & \\ \end{array}$$

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where  $R_{11}$  is hydrogen, O, OH,  $C_1$ - $C_{18}$  alkyl, -CH<sub>2</sub>CN,  $C_1$ - $C_{18}$  alkoxy,  $C_1$ - $C_{18}$  hydroxyalkoxy,  $C_5$ - $C_{12}$  cycloalkoxy,  $C_5$ - $C_{12}$  hydrocycloalkoxy,  $C_5$ - $C_6$  alkenyl,  $C_1$ - $C_{18}$  alkynyl,  $C_7$ - $C_9$  phenylalkyl, unsubstituted or substituted on the phenyl with 1, 2 or 3  $C_1$ - $C_4$  alkyls, or an aliphatic  $C_1$ - $C_8$  acyl;  $R_{12}$  is hydrogen,  $C_1$ - $C_8$  alkyl, or benzyl;  $R_{13}$ ,  $R_{14}$ ,  $R_{15}$ , and  $R_{16}$  are each independently a  $C_1$ - $C_{18}$  alkyl, benzyl or phenethyl, or optionally  $R_{13}$  and  $R_{14}$ , and/or  $R_{15}$  and  $R_{16}$ , taken together with the carbon which they are attached, form a  $C_5$ - $C_{10}$  cycloalkyl.

Another embodiment of a hindered amine compound is a compound that has formula IV below:

$$H = \begin{bmatrix} X & X_1 & N & N \\ 1 & Z & N & N & N \\ N & Z & N & N & N \end{bmatrix}_n$$

where R<sub>10</sub> is a morpholino, C<sub>1</sub> -C<sub>8</sub> alkylamine, di(C<sub>1</sub> -C<sub>8</sub>) alkylamine, pyrrolidyl, cyclohexylamine or combinations thereof,

X and  $X_1$ , which may be the same or different, and are hydrogen,  $C_1$  – $C_{20}$  alkyl, or a radical of formula III defined above,

R<sub>11</sub> to R<sub>18</sub> are as defined above,

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Z is a straight chained or branched C<sub>1</sub>-C<sub>20</sub> alkylene or a straight chained or branched C<sub>1</sub>-C<sub>20</sub> alkalene chain interrupted by at least oxy, thio, or —N(R<sub>17</sub>)—, where R<sub>17</sub> is hydrogen, C<sub>1</sub>-C<sub>20</sub> alkyl, C<sub>5</sub>-C<sub>10</sub> cycloalkylene C<sub>6</sub>-C<sub>12</sub> arylene, C<sub>8</sub>-C<sub>14</sub> aralkylene or the radical of formula III;

n is an integer greater than 1;

20 and Y is a halogen atom, C<sub>1</sub>-C<sub>8</sub> alkylamine, di(C<sub>1</sub>-C<sub>8</sub>) alkylamine, pyrrolidyl, morpholino, cyclohexylamine, or

where X, X<sub>1</sub>, and Z are as previously defined.

Preferably in the above formula IV, Z is a  $C_2$  to  $C_6$  alkylene,  $R_{10}$  is a morpholino or cyclohexylamine, X and  $X_1$  is the radical of formula III,  $R_{11}$  is hydrogen or methyl,  $R_{12}$  is hydrogen, and  $R_{13}$ ,  $R_{14}$ ,  $R_{15}$  and  $R_{18}$  are methyl.

Another embodiment of a hindered amine is a 2,2,6,6-tetraalkylpiperazinone. One embodiment of a 2,2,6,6-tetraalkylpiperazinone compound is one that contains at least one group of formula IVa:

where R<sub>11</sub> is as defined above.

Examples of suitabl hindered amine compounds include, but are not limited to: 1H-Pyrrole-2,5-dione, 1-octadecyl-, polymer with (1-methylethenyl)benzene and 1-(2,2,6,6tetramethyl-4-piperidinyl)-1H-pyrrole-2,5-dione; piperazinone, 1,1',1"-[1,3,5-triazine-2,4,6triyltris[(cyclohexylimino)-2,1-ethanediyl]]tris[3,3,5,5-tetramethyl-]; piperazinone, 1,1',1"-[1,3,5-triazine-2,4,6-triyltris[(cyclohexylimino)-2,1-ethanediyl]]tris[3,3,4,5,5-pentamethyl-]; 5 the reaction product of 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-3,8-diaza-4oxospiro[4.5]decane and epichlorohydrin; the condensate of N,N'-bis(2,2,6,6tetramethylpiperidin-4-yl)hexamethylenediamine and 4-cyclohexylamino-2,6-dichloro-1,3,5triazine; the condensate of 1,2-bis(3-aminopropylamino)ethane; 2,4,6-trichloro-1,3,5-triazine and 4-butylamino-2,2,6,6-tetramethylpiperidine; the condensate of N,N'-bis(2,2,6,6-10 tetramethylpiperidin-4-yl)hexamethylenediamine and 4-morpholino-2,6-dichloro-1,3,5triazine; the condensate of 2-chloro-4,6-bis(4-n-butylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane; the condensate of 2-chloro-4,6bis(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis-(3aminopropylamino)ethane; 2-[(2-hydroxyethyl)amino]-4,6-bis[N-(1-cyclohexyloxy-2,2,6,6-15 tetramethylpiperidin-4-yl)butylamino-1,3,5-triazine; propanedioic acid, [(4-methoxyphenyl)methylene]-bis-(1,2,2,6,6-pentamethyl-4-piperidinyl) ester; tetrakis(2,2,6,6tetramethylpiperidin-4-yl)-1,2,3,4-butanetetracarboxylate; benzenepropanoic acid, 3,5bis(1,1-dimethylethyl)-4-hydroxy-, 1-[2-[3-[3,5-bis(1,1-dimethylethyl)-4-hydroxyphenyl]-1oxopropoxy]ethyl]-2,2,6,6-tetramethyl-4-piperidinyl ester; N-(1-octyloxy-2,2,6,6-20 tetramethylpiperidin-4-yl)-N'-dodecyloxalamide; tris(2,2,6,6-tetramethylpiperidin-4-yl) nitrilotriacetate; 1,5-dioxaspiro{5,5}undecane-3,3-dicarboxylic acid, bis(1,2,2,6,6pentamethyl-4-piperidinyl); 1,5-dioxaspiro{5,5}undecane-3,3-dicarboxylic acid, bis(2,2,6,6tetramethyl-4-piperidinyl); the condensate of 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4hydroxypiperidine and succinic acid; the condensate of N,N'-bis(2,2,6,6-25 tetramethylpiperidin-4-yl)hexamethylenediamine and 4-tert-octylamino-2,6-dichloro-1,3,5triazine; 1,2,3,4-butanetetracarboxylic acid, 1,2,2,6,6-pentamethyl-4-piperidinyl tridecyl ester; tetrakis(2,2,6,6-tetramethylpiperidin-4-yl)-1,2,3,4-butanetetracarboxylate; 1,2,3,4butanetetracarboxylic acid, 2,2,6,6-tetramethyl-4-piperidinyl tridecyl ester; tetrakis(1,2,2,6,6pentamethylpiperidin-4-yl)-1,2,3,4-butanetetracarboxylate; mixture of 2,2,4,4-tetramethyl-30 21-oxo-7-oxa-3.20-diazaspiro(5.1.11.2)-heneicosane-20-propanoic acid-dodecylester; 2,2,4,4-tetramethyl-21-oxo-7-oxa-3.20-diazaspiro(5.1.11.2)-heneicosane-20-propanoic acidtetradecylester; 1H,4H,5H,8H-2,3a,4a,6,7a,8a-hexaazacyclopenta[def]fluorene-4,8-dione, hexahydro-2,6-bis(2,2,6,6-tetramethyl-4-piperidinyl)-; polymethyl[propyl-3-oxy(2',2',6',6'tetramethyl-4,4'-piperidinyl)]siloxane; polymethyl[propyl-3-oxy(1',2',2',6',6'-pentamethyl-4,4'-35 piperidinyl)]siloxane; copolymer of methylmethacrylate with ethyl acrylat and 2,2,6,6-

tetramethylpiperidin-4-yl acrylate; copolymer of mixed C<sub>20</sub> to C<sub>24</sub> alpha-olefins and (2,2,6,6tetramethylpiperidin-4-yl)succinimide; 1,2,3,4-butanetetracarboxylic acid, polymer with β,β,β',β'-tetramethyl-2,4,8,10-tetraoxaspiro[5.5]undecane-3,9-diethanol, 1,2,2,6,6pentamethyl-4-piperidinyl ester, 1,2,3,4-butanetetracarboxylic acid, polymer with β,β,β',β'tetramethyl-2,4,8,10-tetraoxaspiro[5.5]undecane-3,9-diethanol, 2,2,6,6-tetramethyl-4-5 piperidinyl ester copolymer; 1,3-benzenedicarboxamide, N,N'-bis(2,2,6,6-tetramethyl-4piperidinyl; 1,1'-(1,10-dioxo-1,10-decanediyl)-bis(hexahydro-2,2,4.4.6pentamethylpyrimIdine; ethane diamide, N-(1-acetyl-2,2,6,6-tetramethylpiperidinyl)-N'dodecyl; formamide, N,N'-1,6-hexanediylbis[N-(2,2,6,6-tetramethyl-4-piperidinyl); D-glucitol, 10 1.3:2.4-bis-O-(2,2,6,6-tetramethyl-4-piperidinylidene)-; 2,2,4,4-tetramethyl-7-oxa-3,20-diaza-21-oxo-dispiro[5.1.11.2]heneicosane; propanamide, 2-methyl-N-(2,2,6,6-tetramethyl-4piperidinyl)-2-[(2,2,6,6-tetramethyl-4-piperidinyl)amino]-; 7-oxa-3,20diazadispiro[5.1.11.2]heneicosane-20-propanoic acid, 2,2,4,4-tetramethyl-21-oxo-, dodecyl ester; N-(2,2,6,6-tetramethylpiperidin-4-yl)-β-aminopropionic acid dodecyl ester; N-(2,2,6,6-15 tetramethylpiperidin-4-yl)-N'-aminooxalamide; propanamide, N-(2,2,6,6-tetramethyl-4piperidinyl)-3-[(2,2,6,6-tetramethyl-4-piperidinyl)amino]-; mixture of 4-hexadecyloxy- and 4stearyloxy-2,2,6,6-tetramethylpiperidine; 3-dodecyl-1-(1,2,2,6,6-pentamethylpiperidin-4yl)pyrrolidine-2,5-dione; 3-dodecyl-1-(1-ethanoyl-2,2,6,6-pentamethylpiperidin-4yl)pyrrolidine-2,5-dione; bis(2,2,6,6-tetramethylpiperidin-4-yl)succinate; bis(1,2,2,6,6-20 pentamethylpiperidin-4-yl) n-butyl 3,5-di-tert-butyl-4-hydroxybenzylmalonate; tris(2,2,6,6tetramethylpiperidin-4-yl) nitrilotriacetate; 1,1'-(1,2-ethanediyl)bis(3,3,5,5tetramethylpiperazinone); 4-benzoyl-2,2,6,6-tetramethylpiperidine; 4-stearyloxy-2,2,6,6tetramethylpiperidine; bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-tertbutylbenzyl)malonate; 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decan-2,4-dione; bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)sebacate; bis(1-octyloxy-2,2,6,6-25 tetramethylpiperidyl)succinate: 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8triazaspiro[4.5]decane-2.4-dione; 3-dodecyl-1-(2,2,6,6-tetramethylpiperidin-4-yl)pyrrolidin-2,5-dione; 3-dodecyl-1-(1-ethanoyl-2,2,6,6-tetramethylpiperidin-4-yl)pyrrolidin-2,5-dione; 3dodecyl-1-(1,2,2,6,6-pentamethylpiperidin-4-yl)pyrrolidine-2,5-dione; a mixture of 4-30 hexadecyloxy- and 4-stearyloxy-2,2,6,6-tetramethylpiperidine; 2-undecyl-7,7,9,9tetramethyl-1-oxa-3,8-diaza-4-oxospiro[4.5]decane; 1,5-dioxaspiro[5,5]undecane-3,3dicarboxylic acid, bis(2,2,6,6-tetramethyl-4-piperidinyl) and 1,5-dioxaspiro(5,5)undecane-3.3-dicarboxylic acid, bis(1,2,2,6,6-pentamethyl-4-piperidinyl).

The hydroxybenzophenone compound of the present invention may be any suitable hindered hydroxybenzoate compound such as those having the formula V:

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where  $R_{17}$ ,  $R_{18}$  and  $R_{24}$  are each independently hydrogen alkyl, aryl, aralkyl, alkaryl, alkoxy, aryloxy having from 1 to 18 carbon atoms and  $R_{19}$  is hydrogen, a  $C_1$ - $C_{24}$  alkyl, or substituted or unsubstituted  $C_6$ - $C_{24}$  aryl. Preferably,  $R_{17}$ ,  $R_{18}$  and  $R_{24}$  are each hydrogen and  $R_{19}$  is a  $C_1$ - $C_8$  alkyl.

Examples of suitable hindered hydroxybenzophenone compounds include: 2,4-dihydroxybenzophenone; 2-hydroxy-4-methoxybenzophenone; 2-hydroxy-4-octyloxybenzophenone; 2-hydroxy-4-decyloxybenzophenone; 2-hydroxy-4-decyloxybenzophenone; 2,2'-dihydroxy-4-dimethoxybenzophenone; 2,2'-dihydroxy-4-benzyloxybenzophenone; 2,4-dihydroxy-4'-tert-butyl-benzophenone; 1,6-bis-(3-hydroxy-4-benzoylphenoxy)hexane; methylenebis-(2-benzoyl-5-methoxyphenol).

In one embodiment of the present invention, the UV stabilizing additive composition of the present invention may be employed to stabilize materials which are subject to degradation by ultraviolet radiation by contacting the UV stabilizing additive composition with a composition comprising polymeric or other materials, either chemically or physically. Non-limiting examples of materials that may be so stabilized are polyolefins, polyesters, polyethers, polyketones, polyamides, natural and synthetic rubbers, polyurethanes, polystyrenes, high-impact polystyrenes, polyacrylates, polymethacrylates, polyacetals, polyacrylonitriles, polybutadienes, polystyrenes, acrylonitrile-butadiene-styrene, styrene acrylonitrile, acrylate styrene acrylonitrile, cellulosic acetate butyrate, cellulosic polymers, polyimides, polyamideimides, polyetherimides, polyphenylsulfides, polyphenyloxide, polysulfones, polyethersulfones, polyvinylchlorides, polycarbonates, polyketones, aliphatic polyketones, thermoplastic olefins, aminoresin cross-linked polyacrylates and polyesters, polyisocyanate crosslinked polyesters and polyacrylates, phenol/formaldehyde, urea/formaldehyde and melamine/formaldehyde resins, drying and non-drying alkyd resins, alkyd resins, polyester resins, acrylate resins cross-linked with melamine resins, urea resins, isocyanates, isocyanurates, carbamates, and epoxy resins, cross-linked epoxy resins

derived from aliphatic, cycloaliphatic, heterocyclic and aromatic glycidyl compounds, which are cross-linked with anhydrides or amines, polysiloxanes, Michael addition polymers, amines, blocked amines with activated unsaturated and methylene compounds, ketimines with activated unsaturated and methylene compounds, polyketimines in combination with unsaturated acrylic polyacetoacetate resins, polyketimines in combination with unsaturated acrylic resins, radiation curable compositions, epoxymelamine resins, organic dyes, cosmetic products, cellulose-based paper formulations, photographic film paper, fibers, waxes, inks, and blends thereof.

Preferably, the materials to be stabilized are thermoplastic olefins, acrylonitrile-butadiene-styrene, polyesters, polyvinylchloride, polyamides, polyurethanes, or homo- and copolymers of propylene, isobutylene, butene, methylpentene, hexene, heptene, octene, isoprene, butadiene, hexadiene, dicyclopentadiene, ethylidene cyclopentene and norbornene. More preferably, the materials are polypropylene and thermoplastic olefins.

The amount of the triazine compound used in the material to be stabilized using the combination of additives of the present invention is typically lower than normal usage. The lower limit of the amount of triazine may be as low as about 10 ppm, or about 20 ppm, or about 50 ppm or about 75 ppm or about 100 ppm or about 200 ppm, based on the amount of material to be stabilized. There is no upper limit to the amount of triazine, but it would be about 5000 ppm or about 4000 ppm or about 3000 ppm or about 2000 ppm or about 1000 ppm or about 500 ppm, based on the material to be stabilized.

The amount of the hydroxybenzophenone compound used in the material to be stabilized using the combination of additives of the present invention is also typically lower than normal usage. The lower limit of the amount of hydroxybenzophenone may be as low as about 10 ppm, or about 20 ppm, or about 50 ppm or about 75 ppm or about 100 ppm or about 200 ppm, or about 500 ppm, based on the amount of material to be stabilized. There is no upper limit to the amount of hydroxybenzophenone, but it would be about 5000 ppm or about 4000 ppm or about 3000 ppm or about 2000 ppm or about 1000 ppm or about 500 ppm, based on the material to be stabilized.

The amount of the hindered amine compound used in the material to be stabilized using the combination of additives of the present invention is more or less its typically amount. The lower limit of the amount of hindered amine compound may be about 250 ppm, or about 500 ppm, or about 1000 ppm or about 2000 ppm, based on the amount of material to be stabilized. There is no upper limit to the amount of hindered amine compound, but it would be about 30000 ppm, or about 20000 ppm or about 15000 ppm or about 12500 ppm or about 10000 ppm or about 7500 ppm, or about 5000 ppm, based on the material to be stabilized.

The amount of hindered amine compound with respect to the other UV absorbers is typically greater. The ratio of hindered amine compound to the triazine UV absorber may be as high as about 50:1, or about 25:1, or about 20:1 or about 10:1, or about 7:1 or about 3:1. The ratio of hindered amine compound to the hydroxybenzophenone UV absorber may be about 25:1, or about 20:1 or about 10:1, or about 3:1 or about 2:1 or about 1.5:1. The ratio of hydroxybenzophenone compound to the triazine UV absorber may be about 10:1, or about 5:1, or about 3:1 or about 2:1, or about 1:2.

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It should be noted that the amounts and ratios disclosed for the above additives are each independent of each other.

This invention contemplates that a benzotriazole UV absorber may be substituted for the hydroxybenzophenone UV absorber. The benzotriazole compound may also be added to the other three additives. The same ratios and amounts disclosed above for the hydroxybenzophenone UV absorber may be used for the benzotriazole UV absorber. The following are examples of benzotriazole UV absorbers: 2-(2-hydroxy-5-methylphenyl)benzotriazole; 2-(2-hydroxy-5-tert-butylphenyl)-benzotriazole; 2-(2-hydroxy-3,5-di-tertbutylphenyl)-benzotriazole; 5-chloro-2-(2-hydroxy-3-tert-butyl-5-methylphenyl)benzotriazole; 5-chloro-2-(2-hydroxy-3,5-di-tert-butyl-5-methylphenyl)-benzotriazole; 2-(2hydroxy-3-tert-phenyl-5-methyl)-benzotriazole; 2-(2-hydroxy-3,5-di-tert-amyl)-benzotriazole; 2-(2-hydroxy-3-sec-butyl-5-tert-butyl)-benzotriazole; 2-(2-hydroxy-4-octyloxy)-benzotriazole; 2-(2-hydroxy-5-tert-octyl)-benzotriazole; 2-[2-hydroxy-3,5-di(a,a-dimethylbenzyl)phenyl]benzotriazole; 2-(2-hydroxy-3-dodecyl-5-methylphenyl)benzotriazole; 2-[2-hydroxy-3-( $\alpha$ , $\alpha$ 'dimethylbenzyl)-5-(1,1,3,3-tetramethylbutyl)phenyl]benzotriazole; 2,2'-methylenebis[4-(1,1,3,3-tetramethylbutyl)-6-benzotriazol-2-ylphenol]; 2,[2-hydroxy-3-(3,4,5,6tetrahydrophthalimidomethyl)-5-methylphenyl]benzotriazole; 2-[3-tert-butyl-5-(2-(2ethylhexyloxycarbonylethyl))-2-hydroxyphenyl]benzotriazole; a mixture of transesterification products of 2-[(3-tert-butyl-5-(2-methoxycarbonylethyl)-2- hydroxyphenyl)benzotriazole with polyethylene glycol of about MW 300; 5-chloro-2-[2-hydroxy-3-tert-butyl-5-(2octyloxycarbonyl)ethylphenyl]benzotriazole.

This invention also contemplates a method of preparing the compositions above by contacting the UV stabilizing additive composition with the material to be stabilized. The material to be stabilized and UV stabilizing additive composition are contacted by preferably blending or compounding the components in a kneading apparatus such as a single or twin screw extruder, Banbury mixer, or hot rollers. The processing parameters and the use of such kneading apparatuses are well known to those skilled in the art.

As would be apparent to those skilled in the art of making plastic materials, in addition to the material to be stabilized and UV stabilizing additive composition, the

composition of the present invintion may include conventional additives including but are not limited to, antioxidants, metal deactivators, hydroxylamines, nitrones, lactones, costabilizers, nucleating agents, clarifying agents, neutralizers, metallic stearates, metal oxides, hydrotalcites, fillers and reinforcing agents, plasticizers, lubricants, emulsifiers, pigments, rheological additives, catalysts, level agents, optical brighteners, flame retardant agents, anti-static agents and blowing agents.

The UV stabilizing additive composition and the material to be stabilized may be used to make articles, such as an extruded or molded articles, coatings, tapes and films. The articles may be formed by extrusion, sheet extrusion, injection molding, blow molding, injection blow molding, rotational or roto-molding, calendering, thermoforming, compression molding, vacuum molding, pressure molding, reaction injection molding, and other similar techniques known in the art. In addition, coatings may be applied by powder coating, extrusion coating, electrocoating, spraying, dipping, and other similar techniques known in the art.

#### What is claimed is:

- 1. A composition comprising an UV stabilizing composition comprising:
  - (i) an ortho-hydroxy tris-aryl-s-triazine compound;
  - (ii) a hindered amine compound; and
  - (iii) a hydroxybenzophenone compound.
- A composition comprising an UV stabilizing composition comprising:
  - (i) an ortho-hydroxy tris-aryl-s-triazine compound;
  - (ii) a hindered amine compound; and
  - (iii) a hydroxybenzophenone compound

where the ratio of the hindered amine compound to the triazine compound is about 3:1 to about 25:1; and the ratio of the hindered amine compound to the hydroxybenzophenone compound is about 2:1 to 10:1

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- The composition of claim 2 further comprising a material to be stabilized.
- 4. The composition of claim 3 wherein the amount of the triazine compound is about 20 ppm to about 2000 ppm, the amount of the hydroxybenzophenone is about 20 ppm to about 5000 ppm and the amount of the hindered amine is about 250 ppm to about 20000 ppm, all based on the weight of the material to be stabilized.
- from the group consisting of: polyolefins, polyesters, polyethers, polyketones, polyamides, natural and synthetic rubbers, polyurethanes, polystyrenes, high-impact polystyrenes, polyacrylates, polymethacrylates, polyacetals, polyacrylonitriles, polybutadienes, polystyrenes, acrylonitrile-butadiene-styrene, styrene acrylonitrile, acrylate styrene acrylonitrile, cellulosic acetate butyrate, cellulosic polymers, polyimides, polyamideimides, polyetherimides, polyphenylsulfides, polyphenyloxide, polysulfones, polyethersulfones, polyvinylchlorides, polycarbonates, polyketones, aliphatic polyketones, thermoplastic olefins, aminoresin cross-linked polyacrylates and polyesters, polyisocyanate cross-linked polyesters and polyacrylates, phenol/formaldehyde, urea/formaldehyde and melamine/formaldehyde resins, drying and non-drying alkyd resins, alkyd resins, polyester resins, acrylate resins cross-linked with melamine resins, urea resins, isocyanates, isocyanurates, carbamates, and epoxy resins, cross-linked epoxy resins derived from aliphatic, cycloaliphatic, heterocyclic and aromatic glycidyl

compounds, which are cross-linked with anhydrides or amines, polysiloxanes, Michael addition polymers, amines, blocked amines with activated unsaturated and methylene compounds, ketimines with activated unsaturated and methylene compounds, polyketimines in combination with unsaturated acrylic polyacetoacetate resins, polyketimines in combination with unsaturated acrylic resins, radiation curable compositions, epoxymetamine resins, organic dyes, cosmetic products, cellulose-based paper formulations, photographic film paper, fibers, waxes, inks, and blends thereof.

The composition of claim 2, wherein said triazine is selected from the group 6. consisting of: 2,4,6-tris(2-hydroxy-4-octyloxyphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-n-10 octyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-(mixed isooctyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2,4-dihydroxyphenyl)-4,6bis(2,4-dimethylphenyl)-1,3,5-triazine; 2,4-bis(2-hydroxy-4-propyloxyphenyl)-6-(2,4dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-octyloxyphenyl)-4,6-bis(4-methylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-dodecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 15 2-(2-hydroxy-4-tridecyloxyphenyl)-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[2-hydroxy-4-(2-hydroxy-3-butyloxypropyloxy)phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-[2hydroxy-4-(2-hydroxy-3-octyloxypropyloxy)-phenyl]-4,6-bis(2,4-dimethylphenyl)-1,3,5triazine; 2-[4-dodecyloxy/tridecyloxy-2-hydroxypropoxy)-2-hydroxyphenyl]-4,6-bis(2,4dimethylphenyl)-1,3,5-triazine; 2-[2-hydroxy-4-(2-hydroxy-3-dodecyloxypropoxy)phenyl]-4,6-20 bis(2,4-dimethylphenyl)-1,3,5-triazine; 2-(2-hydroxy-4-hexyloxy)phenyl-4,6-diphenyl-1,3,5triazine; 2-(2-hydroxy-4-methoxyphenyl)-4,6-diphenyl-1,3,5-triazine; 2,4,6-tris[2-hydroxy-4-(3-butoxy-2-hydroxypropoxy)phenyl]-1,3,5-triazine; 2-(2-hydroxyphenyl)-4-(4methoxyphenyl)-6-phenyl-1,3,5-triazine and mixtures thereof.

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7. The composition of claim 2, wherein said hindered amine compound is selected from the group consisting of: 1H-Pyrrole-2,5-dione, 1-octadecyl-, polymer with (1-methylethenyl)benzene and 1-(2,2,6,6-tetramethyl-4-piperidinyl)-1H-pyrrole-2,5-dione; piperazinone, 1,1',1"-[1,3,5-triazine-2,4,6-triyltris[(cyclohexylimino)-2,1-ethanediyl]]tris[3,3,5,5-tetramethyl-]; piperazinone, 1,1',1"-[1,3,5-triazine-2,4,6-triyltris[(cyclohexylimino)-2,1-ethanediyl]]tris[3,3,4,5,5-pentamethyl-]; the reaction product of 7,7,9,9-tetramethyl-2-cycloundecyl-1-oxa-3,8-diaza-4-oxospiro[4.5]decane and epichlorohydrin; the condensate of N,N'-bis(2,2,6,6-tetramethylpiperidin-4-yl)hexamethylenediamine and 4-cyclohexylamino-2,6-dichloro-1,3,5-triazine; the condensate of 1,2-bis(3-aminopropylamino)ethane, 2,4,6-trichloro-1,3,5-triazine and 4-butylamino-2,2,6,6-tetramethylpiperidine; the condensate of N,N'-bis(2,2,6,6-

tetramethylpiperidin-4-yl)hexamethylenediamine and 4-morpholino-2,6-dichloro-1,3,5triazin ; the condensate of 2-chloro-4,6-bis(4-n-butylamino-2,2,6,6-tetramethylpiperidyl)-1,3,5-triazine and 1,2-bis(3-aminopropylamino)ethane; the condensate of 2-chloro-4,6bis(4-n-butylamino-1,2,2,6,6-pentamethylpiperidyl)-1,3,5-triazine and 1,2-bis-(3aminopropylamino)ethane; 2-[(2-hydroxyethyl)amino]-4,6-bis[N-(1-cyclohexyloxy-2,2,6,6-5 tetramethylpiperidin-4-yl)butylamino-1,3,5-triazine; propanedioic acid, [(4-methoxyphenyl)methylene]-bis-(1,2,2,6,6-pentamethyl-4-piperidinyl) ester; tetrakis(2,2,6,6tetramethylpiperidin-4-yl)-1,2,3,4-butanetetracarboxylate; benzenepropanoic acid, 3,5bis(1,1-dimethylethyl)-4-hydroxy-, 1-[2-[3-[3,5-bis(1,1-dimethylethyl)-4-hydroxyphenyl]-1oxopropoxy]ethyl]-2,2,6,6-tetramethyl-4-piperidinyl ester; N-(1-octyloxy-2,2,6,6-10 tetramethylpiperidin-4-yl)-N'-dodecyloxalamide; tris(2,2,6,6-tetramethylpiperidin-4-yl) nitrilotriacetate; 1,5-dioxaspiro{5,5}undecane-3,3-dicarboxylic acid, bis(1,2,2,6,6pentamethyl-4-piperidinyl); 1,5-dioxaspiro{5,5}undecane-3,3-dicarboxylic acid, bis(2,2,6,6tetramethyl-4-piperidinyl); the condensate of 1-(2-hydroxyethyl)-2,2,6,6-tetramethyl-4hydroxypiperidine and succinic acid; the condensate of N,N'-bis(2,2,6,6-15 tetramethylpiperidin-4-yl)hexamethylenediamine and 4-tert-octylamino-2,6-dichloro-1,3,5triazine; 1,2,3,4-butanetetracarboxylic acid, 1,2,2,6,6-pentamethyl-4-piperidinyl tridecyl ester; tetrakis(2,2,6,6-tetramethylpiperidin-4-yl)-1,2,3,4-butanetetracarboxylate; 1,2,3,4butanetetracarboxylic acid, 2,2,6,6-tetramethyl-4-piperidinyl tridecyl ester; tetrakis(1,2,2,6,6pentamethylpiperidin-4-yl)-1,2,3,4-butanetetracarboxylate; mixture of 2,2,4,4-tetramethyl-20 21-oxo-7-oxa-3.20-diazaspiro(5.1.11.2)-heneicosane-20-propanoic acid-dodecylester; 2,2,4,4-tetramethyl-21-oxo-7-oxa-3.20-diazaspiro(5.1.11.2)-heneicosane-20-propanoic acidtetradecylester; 1H,4H,5H,8H-2,3a,4a,6,7a,8a-hexaazacyclopenta[def]fluorene-4,8-dione, hexahydro-2,6-bis(2,2,6,6-tetramethyl-4-piperidinyl)-; polymethyl[propyl-3-oxy(2',2',6',6'tetramethyl-4,4'-piperidinyl)]siloxane; polymethyl[propyl-3-oxy(1',2',2',6',6'-pentamethyl-4,4'-25 piperidinyl)]siloxane; copolymer of methylmethacrylate with ethyl acrylate and 2,2,6,6tetramethylpiperidin-4-yl acrylate; copolymer of mixed C20 to C24 alpha-olefins and (2,2,6,6tetramethylpiperidin-4-yl)succinimide; 1,2,3,4-butanetetracarboxylic acid, polymer with  $\beta,\beta,\beta',\beta'$ -tetramethyl-2,4,8,10-tetraoxaspiro[5.5]undecane-3,9-diethanol, 1,2,2,6,6pentamethyl-4-piperidinyl ester; 1,2,3,4-butanetetracarboxylic acid, polymer with  $\beta,\beta,\beta',\beta'$ -30 tetramethyl-2,4,8,10-tetraoxaspiro[5.5]undecane-3,9-diethanol, 2,2,6,6-tetramethyl-4piperidinyl ester copolymer; 1,3-benzenedicarboxamide, N,N'-bis(2,2,6,6-tetramethyl-4piperidinyl; 1,1'-(1,10-dioxo-1,10-decanediyl)-bis(hexahydro-2,2,4,4.6pentamethylpyrimidine; ethane diamide, N-(1-acetyl-2,2,6,6-tetramethylpiperidinyl)-N'dodecyl; formamide, N,N'-1,6-hexanediylbis[N-(2,2,6,6-tetramethyl-4-piperidinyl); D-glucitol, 35 1,3:2,4-bis-O-(2,2,6,6-tetramethyl-4-piperidinylidene)-; 2,2,4,4-tetramethyl-7-oxa-3,20-diaza-

21-oxo-dispiro[5.1.11.2]heneicosane; propanamide, 2-methyl-N-(2,2,6,6-tetramethyl-4piperidinyl)-2-[(2,2,6,6-tetramethyl-4-piperidinyl)amino]-; 7-oxa-3,20diazadispiro[5.1.11.2]heneicosane-20-propanoic acid, 2,2,4,4-tetramethyl-21-oxo-, dodecyl ester; N-(2,2,6,6-tetramethylpiperidin-4-yl)-β-aminopropionic acid dodecyl ester; N-(2,2,6,6tetramethylpiperidin-4-yl)-N'-aminooxalamide; propanamide, N-(2,2,6,6-tetramethyl-4-5 piperidinyl)-3-[(2,2,6,6-tetramethyl-4-piperidinyl)amino]-; mixture of 4-hexadecyloxy- and 4stearyloxy-2,2,6,6-tetramethylpiperidine; 3-dodecyl-1-(1,2,2,6,6-pentamethylpiperidin-4yl)pyrrolidine-2,5-dione; 3-dodecyl-1-(1-ethanoyl-2,2,6,6-pentamethylpiperidin-4yl)pyrrolidine-2,5-dione; bis(2,2,6,6-tetramethylpiperidin-4-yl)succinate; bis(1,2,2,6,6pentamethylpiperidin-4-yl) n-butyl 3,5-di-tert-butyl-4-hydroxybenzylmalonate; tris(2,2,6,6-10 tetramethylpiperidin-4-yl) nitrilotriacetate; 1,1'-(1,2-ethanediyl)bis(3,3,5,5tetramethylpiperazinone); 4-benzoyl-2,2,6,6-tetramethylpiperidine; 4-stearyloxy-2,2,6,6tetramethylpiperidine; bis(1,2,2,6,6-pentamethylpiperidyl)-2-n-butyl-2-(2-hydroxy-3,5-di-tertbutylbenzyl)malonate; 3-n-octyl-7,7,9,9-tetramethyl-1,3,8-triazaspiro[4.5]decan-2,4-dione; bis(1-octyloxy-2,2,6,6-tetramethylpiperidyl)sebacate; bis(1-octyloxy-2,2,6,6-15 tetramethylpiperidyl)succinate; 8-acetyl-3-dodecyl-7,7,9,9-tetramethyl-1,3,8triazaspiro[4.5]decane-2,4-dione; 3-dodecyl-1-(2,2,6,6-tetramethylpiperidin-4-yl)pyrrolidin-2.5-dione; 3-dodecyl-1-(1-ethanoyl-2,2,6,6-tetramethylpiperidin-4-yl)pyrrolidin-2,5-dione; 3dodecyl-1-(1,2,2,6,6-pentamethylpiperidin-4-yl)pyrrolidine-2,5-dione; a mixture of 4hexadecyloxy- and 4-stearyloxy-2,2,6,6-tetramethylpiperidine; 2-undecyl-7,7,9.9-20 tetramethyl-1-oxa-3,8-diaza-4-oxospiro[4.5]decane; 1,5-dioxaspiro{5,5}undecane-3,3dicarboxylic acid, bis(2,2,6,6-tetramethyl-4-piperidinyl) and 1,5-dioxaspiro(5,5)undecane-3,3-dicarboxylic acid, bis(1,2,2,6,6-pentamethyl-4-piperidinyl) and mixtures thereof.

- 8. The composition of claim 2 wherein said hydroxybenzophenone is selected from the group consisting of 2,4-dihydroxybenzophenone, 2-hydroxy-4-methoxybenzophenone, 2-hydroxy-4-decyloxybenzophenone, 2-hydroxy-4-decyloxybenzophenone, 2-hydroxy-4-decyloxybenzophenone, 2-hydroxy-4-decyloxybenzophenone, 2,2'-dihydroxy-4,4'-dimethoxybenzophenone, 2,2',4,4'-tetrahydroxybenzophenone, 2,4-dihydroxy-4'-tert-butyl-benzophenone; 1,6-bis-(3-hydroxy-4-benzoylphenoxy)hexane; methylenebis-(2-benzoyl-5-methoxyphenol) and mixtures thereof.
  - 9. The composition of claim 2, further comprising a benzotriazole compound.
- 35 10. A composition comprising an UV stabilizing composition comprising:
  - (i) an ortho-hydroxy tris-aryl-s-triazine compound;

- (ii) a hindered amine compound; and
- (iii) a benzotriazole compound

wherein the ratio of the hindered amine compound to the triazine compound is about 3:1 to about 25:1; and the ratio of the hindered amine compound to the benzotriazole compound is about 2:1 to 10:1.

- 11. The composition of claim 10 further comprising a material to be stabilized.
- 12. The composition of claim 11 wherein the amount of the triazine compound is about 20 ppm to about 2000 ppm, the amount of the benzotriazole is about 20 ppm to about 5000 ppm and the amount of the hindered amine is about 250 ppm to about 20000 ppm, all based on the weight of the material to be stabilized.
- The composition of claim 10, wherein the benzotriazole compound is selected from 13. the group consisting of: 2-(2-hydroxy-5-methylphenyl)-benzotriazole, 2-(2-hydroxy-5-tert-15 butylphenyl)-benzotriazole, 2-(2-hydroxy-3,5-di-tert-butylphenyl)-benzotriazole, 5-chloro-2-(2-hydroxy-3-tert-butyl-5-methylphenyl)-benzotriazole, 5-chloro-2-(2-hydroxy-3,5-di-tertbutyl-5-methylphenyl)-benzotriazole, 2-(2-hydroxy-3-tert-phenyl-5-methyl)-benzotriazole, 2-(2-hydroxy-3,5-di-tert-amyl)-benzotriazole, 2-(2-hydroxy-3-sec-butyl-5-tert-butyl)benzotriazole, 2-(2-hydroxy-4-octyloxy)-benzotriazole, 2-(2-hydroxy-5-tert-octyl)-20  $benzotriazole,\ 2\hbox{-}[2\hbox{-hydroxy-3,5-di}(\alpha,\alpha\hbox{-dimethylbenzyl}) phenyl]\hbox{-benzotriazole};\ 2\hbox{-}(2\hbox{-hydroxy-dimethylbenzyl})$  $\textbf{3-dodecyl-5-methylphenyl)} benzotriazole; \textbf{2-[2-hydroxy-3-(}\alpha,\alpha'-dimethylbenzyl)-5-(1,1,3,3-dodecyl-5-methylphenyl)} \\$ tetramethylbutyl)phenyl]benzotriazole; 2,2'-methylenebis[4-(1,1,3,3-tetramethylbutyl)-6benzotriazol-2-ylphenol]; 2,[2-hydroxy-3-(3,4,5,6-tetrahydrophthalimidomethyl)-5methylphenyl]benzotriazole; 2-[3-tert-butyl-5-(2-(2-ethylhexyloxycarbonylethyl))-2-25 hydroxyphenyl]benzotriazole; a mixture of transesterification products of 2-[(3-tert-butyl-5-(2-methoxycarbonylethyl)-2-hydroxyphenyl)benzotriazole with polyethylene glycol of about MW 300; 5-chloro-2-[2-hydroxy-3-tert-butyl-5-(2-octyloxycarbonyl)ethylphenyl]benzotriazole and mixtures thereof.

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